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First-principle dynamical electronic characteristics of Al electromigration in the bulk, surface, and grain boundary

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Absence of density of states transfer observed by interlayer tunneling spectroscopy in magnetic fields for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$

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Using short-pulse interlayer tunneling spectroscopy, the superconducting gap and the pseudogap structure are measured both in the absence and presence of magnetic fields for slightly overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$. It is found, together with characteristic field-dependent behavior of the superconducting conductance peak, that the tunneling conductance within the superconducting gap remains almost unchanged under magnetic fields up to 9 T. This implies that the transfer of the quasiparticle density of states associated with the superconducting transition is absent in this system, suggesting unconventional pairing interaction or a field-induced ordered state that competes with superconductivity.

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It is probably true that the understanding of the pseudogap¹ provides a key to the mechanism of high- T_c superconductivity in cuprates. Although various models to explain the pseudogap have been proposed,^{2–8} a general consensus is yet to be reached. In order to further reveal the nature of the pseudogap and its relation to the superconducting gap, it is imperative to measure these gap structures in the presence of magnetic fields B . Even for high- T_c superconductors, a high magnetic field suppresses superconductivity substantially in a more or less limited temperature (T) range below T_c . This enables the observation of the temperature evolution, if any, of the pseudogap in an extended T range. Furthermore, the application of a magnetic field causes transfer of the quasiparticle density of states (DOS) from above the superconducting gap to energies near the Fermi level. Therefore, the observation of tunneling conductance as a function of magnetic fields provides important information concerning the relationship between the pseudogap and the superconducting gap. And, more importantly, this may lead to a probe into the pairing interaction.

Measurements of the gap structure in the presence of magnetic fields were attempted recently by tunneling spectroscopy for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$.^{9–12} Krasnov *et al.*^{10,11} and others¹² reported that the pseudogap is independent of temperature and magnetic fields for an optimally doped specimen and concluded that the superconducting gap and the pseudogap coexist in the superconducting state. Renner *et al.*⁹ measured the tunneling conductance in the vortex core normal states to draw a conclusion that both gaps are T independent and the pseudogap reflects the incoherent pair states. However, these experiments lacked the observation of quasiparticle DOS transfer associated with the superconducting gap in magnetic fields.

In order to know the pseudogap behavior in detail in an extended T range and to observe the field-induced quasiparticle DOS transfer, we have measured the pseudogap and the superconducting gap in the presence of magnetic fields up to 9 T for slightly overdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ by interlayer

tunneling spectroscopy (ITS).¹³ Among various methods that probe the gap structures in high- T_c superconductors, ITS is a unique method in that it probes the quasiparticle energy spectrum under little influence from vulnerable surface states of specimens. It also attains a high resolution of superconductor/insulator/superconductor (SIS) tunneling spectroscopy. This method utilizes a combination of a short-pulse technique and a small mesa structure comprising several intrinsic tunnel Josephson junctions. Using this method, it is found that the pseudogap peak evolves with decreasing temperature below T_c when the superconductivity is suppressed in the presence of a magnetic field. It is also found that the quasiparticle DOS at the Fermi level ($E=0$) remains almost unchanged even when the magnetic field is increased up to 9 T. This result is striking because it apparently means that the DOS is not conserved in the meaning of the BCS mechanism when the magnetic field is applied. We argue that this is related to a field-induced increase in the degree of order that may originate from spatially inhomogeneous spins and charges, or to an unconventional pairing mechanism.

The specimens used for ITS are $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ mesas with a lateral size of 5 to 10 μm and a thickness of 15 nm. The number of intrinsic tunnel Josephson junctions, N , in these mesas is approximately ten. The thinness and smallness of such a mesa, and a rather thick Au upper electrode layer (400–500 nm), together with a short-pulse method, effectively reduce the self-heating due to current injection during measurements. The mesas were fabricated by engraving a cleaved surface of a $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystal grown by the traveling-solvent–floating-zone method.¹⁴ The details of the fabrication method were described elsewhere.^{13,15} From the temperature dependence of the mesa resistance, which is proportional to the c -axis resistivity ρ_c , we determined the doping level δ by using the empirical relationship between ρ_c and δ .^{14,16} The doping levels for specimens in the present study ranged from $\delta=0.25$ to 0.28 and thus the specimens are in the slightly overdoped region. Although a result of a single specimen is presented below, all the six specimens measured exhibited similar characteristics.

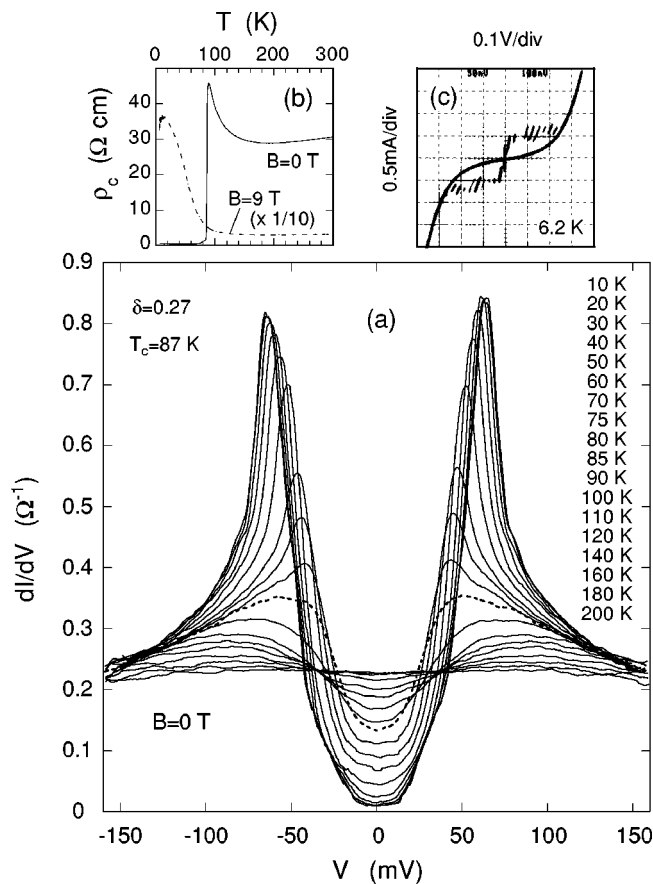


FIG. 1. (a) dI/dV characteristics for $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ with $\delta = 0.27$ in the absence of a magnetic field measured by short-pulse interlayer tunneling spectroscopy. The specimen is a $10 \times 10 \mu\text{m}^2$ mesa with a thickness of 15 nm, which corresponds to ten intrinsic junctions connected in series. The thick dashed curve represents the characteristics at 85 K, close to T_c . (b) Temperature dependence of ρ_c for this mesa. Contact resistance is 0.7Ω , approximately 1% of the mesa resistance at 300 K. (c) Oscilloscope image of I - V characteristics for this mesa at 6.2 K, showing ten resistive branches.

In the short-pulse tunneling method, current pulses with a width of $1.7 \mu\text{s}$ were supplied with an arbitrary wave form generator. Current pulses have a smooth shape with a quarter-period sinusoidal curve both at the rise and fall parts, which serve to improve the breakdown voltage of a mesa. The voltage responses were measured at $0.7 \mu\text{s}$ from the rise of a pulse. Magnetic fields up to 9 T were applied parallel to the c axis with a superconducting magnet.

Figure 1 shows the results for a specimen with $T_c = 87 \text{ K}$, $N = 10$, $\delta = 0.27$, and a size of $10\text{-}\mu\text{m}$ square. $\rho_c(T)$ measured for this mesa is shown in Fig. 1(b). The contact resistance is $\sim 1\%$ of ρ_c (300 K). Figure 1(c) shows an oscilloscope image of I - V characteristics, providing a value of $N = 10$ for the resistive branches. The mesa thickness was determined by this value for N . It is clear that the characteristics are rather homogeneous for all the junctions in the mesa. From this, it is acceptable that the junctions behave similarly in the presence of a magnetic field. Figure 1(a) shows a set of dI/dV - V curves [tunneling conductance, $\sigma_t(V)$] in the absence of magnetic fields for various tem-

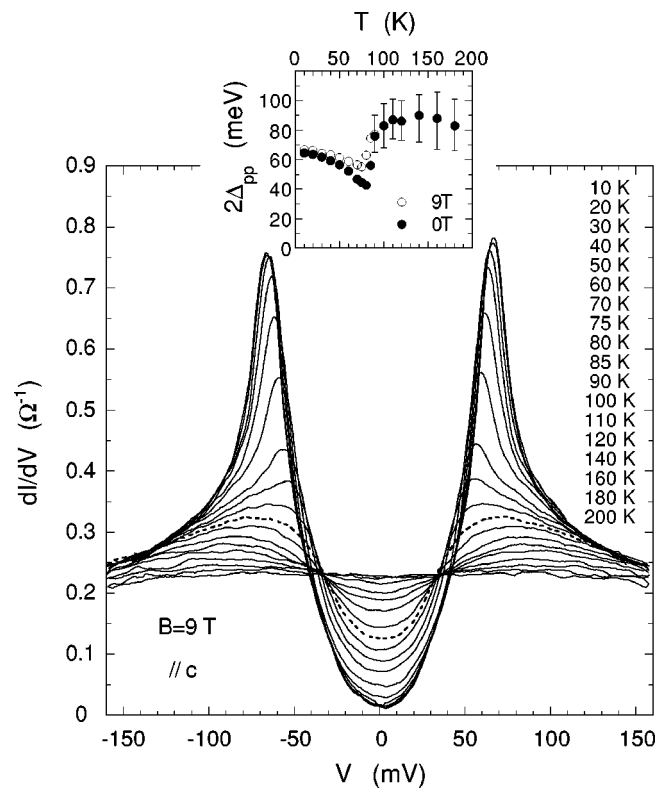


FIG. 2. dI/dV characteristics for the same specimen in Fig. 1 by short-pulse interlayer tunneling spectroscopy in the presence of a magnetic field of 9 T parallel to the c axis. The thick dashed curve represents the characteristics at 85 K, close to T_c . The inset shows the temperature dependence of half the peak separation $2\Delta_{pp}$ at two different fields.

peratures below and above T_c up to 200 K. The voltages V here and hereafter are values normalized by N for a single junction. It is seen that the superconducting gap magnitude $2\Delta_S$, defined as half the peak separation, is 64 meV at 10 K, which is reasonable compared with other spectroscopic results of 60–80 meV.^{17,18} The superconducting conductance peak decreases both in magnitude and peak separation with increasing T . Near T_c , the superconducting peak changes to a round cusp at the shoulder of the broad background, which is the pseudogap, and then disappears at T_c . Above T_c , the broad pseudogap remains and persists up to about 200 K in this specimen. This behavior is consistent with our earlier report for a specimen with a similar doping level and reinforces the argument that the pseudogap and the superconducting gap are different.^{16,19,20}

Figure 2 shows a set of dI/dV - V curves [$\sigma_t(V)$] for the same specimen in the presence of a magnetic field of 9 T. At a first glance, it appears that the overall feature remains almost unchanged except that the shift of the superconducting peak with increasing T becomes less discernible. This indicates that the superconducting peak shifts toward higher energies by the application of magnetic fields. To see this more clearly, we plot half the peak separation $2\Delta_{pp}$ as a function of temperature in the inset to Fig. 2 for both cases of $B = 0 \text{ T}$ and 9 T. This behavior is apparently contradictory to the conventional behavior of the superconducting gap in the

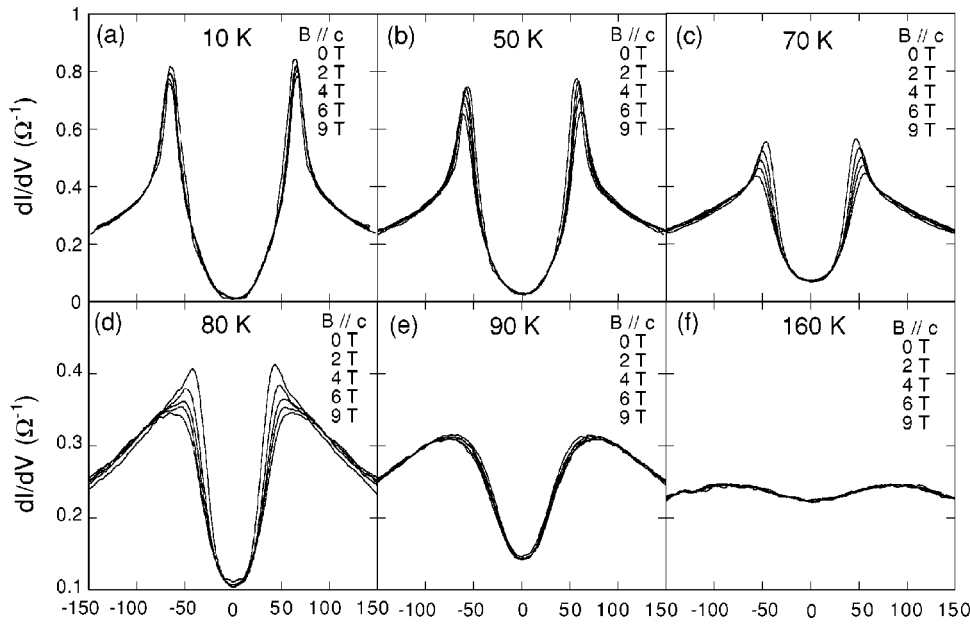


FIG. 3. dI/dV characteristics under different magnetic fields parallel to the c axis at different temperatures for the same specimen in Fig. 1, by interlayer tunneling spectroscopy.

presence of magnetic fields. In the BCS theory for the weak-coupling limit, the magnetic-field effect on the quasiparticle DOS is dealt with in terms of pair breaking.²¹ Within this framework, the superconducting peak in the presence of magnetic fields shows no shift, but rather the peak is broadened as a result of transfer of the quasiparticle DOS from energies $E > \Delta_S$ to $E < \Delta_S$. Such behavior is missing in the result shown in Fig. 2. The present experimental result can be explained in two ways. In the first case, it is postulated that the $\sigma_t(V)$ peak is a composite of the superconducting peak and the pseudogap, and that only the superconducting peak is depressed in the presence of a magnetic field. If the pseudogap peak is located at a slightly higher than Δ_S energy position, then the application of a magnetic field eventually causes the shift of the $\sigma_t(V)$ peak toward higher energies, as argued by Krasnov *et al.*¹⁰ The second explanation invokes the unconventional mechanism of superconductivity which is totally different from the BCS mechanism. However, when we take into account that the superconducting gap and the pseudogap are distinct in the underdoped region,^{16,19} the present result is likely to reflect that the superconducting peak and the pseudogap peak lie in close proximity and behave differently in the presence of magnetic fields.

Figure 3 shows the magnetic-field dependence of $\sigma_t(V)$ at various temperatures from 10 K below T_c to 160 K above T_c . At 10 K, the change caused by magnetic fields is only recognized as a slight decrease in the peak height of $\sigma_t(V)$ and a slight shift of the peak center toward higher energies. It is obvious that the $\sigma_t(V)$ peak is mostly ascribed to the superconductivity because a magnetic field of 9 T is insufficient to bring about an appreciable change in the DOS at 10 K. Since the mean-field upper critical field H_{c2} for the $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ system is estimated to be no higher than 90 T,²² the Ginzburg-Landau (GL) coherence length ξ_{GL} at low temperatures is estimated to be smaller than 1.9 nm. Then it turns out that the normal area due to vortex cores is approximately 5% at 9 T. This value is roughly compared with the change in $\sigma_t(V)$. Taking into account the T depen-

dence of $\xi_{\text{GL}} \propto (1 - T/T_c)^{-0.5}$, we find that this specimen undergoes the mean-field transition to the normal state at approximately 78 K and $B = 9$ T.²³ Therefore, the conductance curves at $B = 9$ T and $T > 80$ K mostly represent an energy-gap structure which is not due to superconductivity. Then, the remaining gap structure is the pseudogap, indicating that the pseudogap exists below T_c .

Between T ranges above and below T_c , there is a qualitative difference in the field-dependent behavior of $\sigma_t(V)$. Above T_c , $\sigma_t(V)$ is almost field independent. A small field-induced change in $\sigma_t(V)$ at 90 K is most probably due to the fluctuation conductivity of the Aslamazov-Larkin type. This implies that the pseudogap is almost field independent at least in a B range of no higher than 9 T.

Another unconventional feature observed in the tunneling spectroscopy under magnetic fields is the almost unchanged $\sigma_t(V)$ below the gap voltage, especially near the Fermi level ($V \approx 0$), as clearly seen in Fig. 3. Strictly speaking, $\sigma_t(V)$ of an SIS junction is different from the quasiparticle DOS. In the present case, however, we treat $\sigma_t(V)$ approximately as a measure of the quasiparticle DOS, because the SIS tunneling conductance approximately reflects the quasiparticle DOS except for the magnitude which displays rather enhanced structure. Thus, the present result reflects a field-induced change in quasiparticle DOS, which highlights the very anomalous feature seen in high- T_c superconducting $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$.

In conventional superconductors, magnetic fields induce transfer of the quasiparticle DOS from the peak near $E = \Delta_S$ to lower energies below Δ_S . For any $E > \Delta_S$, the DOS decreases with increasing B and by no means exceeds the DOS at $B = 0$ T. For $E < \Delta_S$, on the other hand, the DOS increases with increasing B . The DOS at $E = 0$ increases with increasing B up to the DOS value of the normal state for $B > B_{c2}$. In light of this conventional behavior of the magnetic-field-dependent quasiparticle DOS, the magnetic-field dependence seen in Fig. 3 is quite anomalous in that the

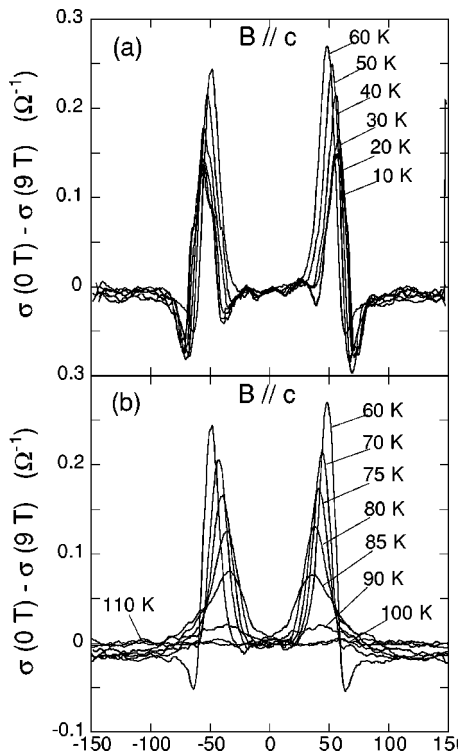


FIG. 4. Subtracted tunneling conductance $\sigma_t(0 \text{ T}) - \sigma_t(9 \text{ T})$ at different temperatures, showing quasiparticle DOS transfer due to the application of a magnetic field of 9 T parallel to the c axis.

quasiparticle DOS at $E=0$ remains almost unchanged. From the principle of the conservation of the density of states, this anomalous result implies that the quasiparticle DOS at the superconducting peak position comes from energies higher than Δ_S and not from the states at $E < \Delta_S$. This implies that the pairing mechanism of high- T_c superconductivity is totally different from the BCS mechanism.

In order to examine the DOS transfer more closely, we plot the difference of $\sigma_t(V)$ for $B=0 \text{ T}$ and $B=9 \text{ T}$ in Figs. 4(a) and 4(b), respectively. Below 60 K, $\Delta\sigma_t(V, 9 \text{ T}) = \sigma_t(V, 0 \text{ T}) - \sigma_t(V, 9 \text{ T})$ increases with T , indicating that the total normal vortex core area at $B=9 \text{ T}$ increases with T .²⁴ Above 60 K, $\Delta\sigma_t(V, 9 \text{ T})$ decreases with T , reflecting that almost all the superfluid density, which decreases with T , is turned normal at $B=9 \text{ T}$. These contrasting T -dependent behaviors of $\Delta\sigma_t(V, 9 \text{ T})$ in the opposite directions imply that superconductivity is substantially suppressed at $B=9 \text{ T}$ above 70 K. From Fig. 4, it is clearly seen that $\Delta\sigma_t(V, 9 \text{ T}) \approx 0$ within the superconducting gap and the DOS transfer is clearly missing. In conventional superconductors, the field-induced difference of the quasiparticle DOS, i.e., $\Delta N(E, B) = N(E, 0 \text{ T}) - N(E, 9 \text{ T})$, or $\Delta\sigma_t(V, 9 \text{ T})$

in the present case, is represented by both a negative dip centered at $E=0$ and the superconducting gap peak centered at $E=\Delta_S$. Compared with this conventional behavior, the present result is decisively different in that $\Delta\sigma_t(V, 9 \text{ T})$ is nearly zero and not negative unlike conventional superconductors. This behavior is striking. It may imply that the mechanism of pairing in high- T_c superconductors is totally different from conventional one.

There are two scenarios that may account for the missing DOS transfer from the superconducting peak to the Fermi energy. The first scenario invokes a totally unconventional pairing mechanism. In the phonon mechanism of pairing, electrons near the Fermi level are involved essentially, while in this scenario, the quasiparticles involved in pairing occupy states quite distant from the Fermi energy. Figure 4(a) shows the negative maximum of $\Delta\sigma_t(V)$ near $V=70 \text{ mV}$, which is located at a slightly larger than $2\Delta_S/e$ position. This negative peak in $\Delta\sigma_t$ might partially account for the DOS transfer to the quasiparticle conductance peak. However, as clearly seen in Fig. 4(a), the area of this negative portion is insufficient to balance the $\Delta\sigma(V)$ peak at $V=2\Delta_S/e$. Therefore, it may be reasoned that the quasiparticle DOS at $V=2\Delta_S/e$ comes from quite distant energy levels. Such behavior may be in line with the kinetic-energy-driven mechanism,^{25–28} in which the kinetic energy lowering at high-energy states gives rise to a superconducting condensate.

Another scenario invokes competition between the superconducting state and another ordered state of a different kind. In this scenario, the unknown ordered state becomes energetically stable in the presence of a magnetic field. As a result of this different ordered state, the quasiparticle DOS near the Fermi level is partly expelled to distant energy levels outside the present tunneling spectroscopy range. This ordered state induced by magnetic field is compared with recent experiments suggesting antiferromagnetic order around the vortex cores.²⁹ In this case, the pseudogap structure in the presence of magnetic fields is considered to manifest itself as an ordered state, which may be related to such an antiferromagnetic state.

In conclusion, we have observed the absence of conventional quasiparticle DOS transfer by interlayer tunneling spectroscopy in the presence of magnetic fields. The result is totally at variance with the conventional pairing mechanism. This is thought to be relevant to an unconventional pairing mechanism such as a kinetic-energy-driven model, or to a magnetic-field-induced antiferromagnetic ordered state that competes with the superconductivity in the presence of a magnetic field.

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